Piezoelectric field-enhanced second-order nonlinear optical susceptibilities in wurtzite GaN/AlGaN quantum wells

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Second-order nonlinear optical processes including second-harmonic generation, optical rectification, and difference-frequency generation associated with intersubband transitions in wurtzite GaN/AlGaN quantum well (QW) are investigated theoretically. Taking into account the strain-induced piezoelectric (PZ) effects, we solve the electronic structure of the QW from coupled effective-mass Schrödinger equation and Poisson equation including the exchange-correlation effect under the local-density approximation. We show that the large PZ field in the QW breaks the symmetry of the confinement potential profile and leads to large second-order susceptibilities. We also show that the interband optical pump-induced electron-hole plasma results in an enhancement in the maximum value of the nonlinear coefficients and a redshift of the peak position in the nonlinear optical spectrum. By use of the difference-frequency generation, THz radiation can be generated from a $GaN/Al_{0.75}Ga_{0.25}N$ with a pump laser of 1.55 μm. © *2000 American Institute of Physics.* [S0003-6951(00)01203-1]

During the past decade there has been considerable interest in the second-order nonlinear optical properties of semiconductor quantum wells (QWs) associated with intersubband transitions.[1–9] In most of the previous work,[1–8] an asymmetric QW structure was used to obtain a large second-order nonlinearity. Because of the small conduction band offset for QW structures such as $GaAs/Al_xGa_{1-x}As$, the second-harmonic generation and optical rectification were usually measured at the wavelength around 10 μm by use of CO_2 lasers. The experimentally used structures include compositionally asymmetric quantum wells,[3,5] asymmetrically coupled quantum wells,[4,6] and applied-field-biased quantum wells.[2]

In this letter we show that the large strain-induced piezoelectric (PZ) field in a wurtzite GaN/AlGaN quantum well breaks the symmetry of the confinement potential profile and leads to large second-order susceptibilities. We also show that the nonlinear susceptibilities can be controlled by an external dc field or an interband optical pump field. Depending on the bias direction relative to the PZ field direction, the applied dc field leads to either a blue- or redshift of the peak position of the nonlinear optical spectrum due to the quantum-confined Stark effect, while the pump-induced electron-hole plasma results in a redshift of the nonlinear spectrum because of the many-body screening effect. Owing to the large band offset, THz radiation can be generated from a $GaN/Al_{0.75}Ga_{0.25}N$ QW by use of difference-frequency generation with a pump laser of 1.55 μm.

We consider a (0001)-grown GaN/AlGaN QW structure in the presence of two optical fields with angular frequencies of ω_1 and ω_2 with the polarization along the well growth direction (z axis). In the second-order perturbation theory, the *zzz* component of the nonlinear susceptibility tensor that describes the sum-frequency generation $(\omega_1+\omega_2)$ and difference-frequency generation $(\omega_1-\omega_2)$ processes is given by[10]

$$\chi^{(2)}(\omega_1\pm\omega_2;\omega_1,\pm\omega_2) = \frac{e^3}{2\epsilon_0}\sum_{l,k,m}\frac{\langle z\rangle_{lk}\langle z\rangle_{km}\langle z\rangle_{ml}}{\hbar(\omega_1\pm\omega_2)-E_{kl}+i\Gamma_{kl}}\Bigg\{(N_l-N_m)\times\left[\frac{1}{\hbar\omega_1-E_{ml}+i\Gamma_{ml}}+\frac{1}{\pm\hbar\omega_2-E_{ml}+i\Gamma_{ml}}\right] + (N_k-N_m)\left[\frac{1}{\hbar\omega_1-E_{km}+i\Gamma_{km}} + \frac{1}{\pm\hbar\omega_2-E_{km}+i\Gamma_{km}}\right]\Bigg\}. \quad (1)$$

In Eq. (1), $\langle z\rangle_{lk}=\int\psi_l^*(z)z\psi_k(z)dz$, $E_{kl}=E_k-E_l$, where E_l and $\psi_l(z)$ are the eigenenergy and the corresponding envelope wave function of the lth subband of the QW. The quantity N_l is the 3D electron density in the lth subband and Γ_{kl} is the line broadening factor for the transition between subbands k and l. Note that we have neglected the conduction band nonparabolicity in deriving Eq. (1) for simplicity.

To obtain the eigenenergies and envelope wave functions of the QW, we solve the effective-mass Schrödinger equation coupled with the Poisson equation. The exchange-correlation effect is included in the local-density approximation.[11] The conduction band offset is given by $\Delta E^c + P_{c\epsilon}$ where ΔE^c is the conduction band discontinuity between the well and barrier layers in the absence of strain and

$$P_{c\epsilon}=2a_c\left(1-\frac{C_{13}}{C_{33}}\right)\epsilon_\parallel \quad (2)$$

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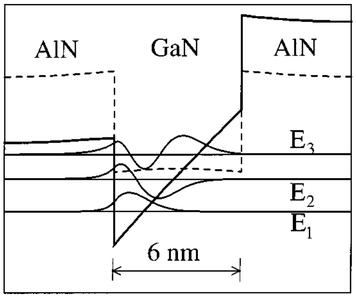

FIG. 1. Self-consistent potential and the three lowest energy levels with corresponding wave functions of a GaN/AlN quantum well with a GaN width of 6 nm. The 5 nm wide barrier layers are doped with an ionized donor concentration of 5×10^{18} cm^{-3}. The self-consistent potential without the piezoelectric field is shown in dashed curve.

is a hydrostatic energy shift due to strain.[12] The strain-induced piezoelectric field is given by[12]

$$F_z^w=\frac{2d_{31}}{\epsilon_0\epsilon_r}\left(C_{11}+C_{12}-\frac{2C_{13}^2}{C_{33}}\right)\epsilon_\parallel, \tag{3}$$

where d_{31} is the piezoelectric constant, ϵ_r is the relative dielectric constant, C_{ij} is the elastic stiffness coefficient, $\epsilon_\parallel=(a_s-a_e)/a_e$, a_s and a_e are the lattice constants of the substrate and the epitaxial layer, respectively. For the GaN/AlGaN QW we take $a_c=-4.08$ eV.[13] For a 6 nm GaN well sandwiched between AlN barrier layers, the strain-induced PZ field is estimated to be about 4.28 MV/cm. The relevant parameters are taken from Ref. 12. Under a modulation doping with an ionized donor concentration of 5×10^{18} cm^{-3}, the self-consistent potential and the three lowest energy wave functions are shown in Fig. 1. For comparison, we also plot the self-consistent potential without the PZ field in the dashed curve. It is clear from Fig. 1 that the large PZ field in the well significantly changes the symmetry of the potential profile of our symmetric QW without an applied field. Thus we would expect large second-order nonlinear coefficients in such a system.

In Fig. 2 we show the second-harmonic ($\chi_{2\omega}^{(2)}$) and optical rectification ($\chi_0^{(2)}$) susceptibilities of a GaN/AlN QW as a function of the fundamental photon energy $\hbar\omega$ for different applied dc fields. Here we consider the applied field is positive when it is along the same direction as the PZ field. We see from Fig. 2 that the second-order nonlinear effects are nonzero for the symmetric QW without the applied field because the strain induced PZ field breaks the symmetry of the system. In the frequency range in Fig. 2(a) there are two resonance peaks in the second-harmonic spectrum. The lower energy peak is due to the resonance at $2\hbar\omega=E_{31}$, while the higher energy peak stems from the resonance at $\hbar\omega=E_{21}$. There is also a resonant peak [not shown in Fig. 2(a)] when the fundamental photon energy is equal to E_{31}. But the magnitude of that peak is smaller than those in Fig. 2(a). For the optical rectification spectrum there are two resonance peaks. One is located at $\hbar\omega=E_{21}$, and the other is at $\hbar\omega=E_{31}$ [not shown in Fig. 2(b)]. When the QW is subjected to an applied field, the quantum-confined Stark effects shift the intersubband transition energies and lead to a shift of the peak in the nonlinear spectra. We also note from Fig. 2 that the peak values of the nonlinear coefficients slightly increase when the QW is negatively biased.

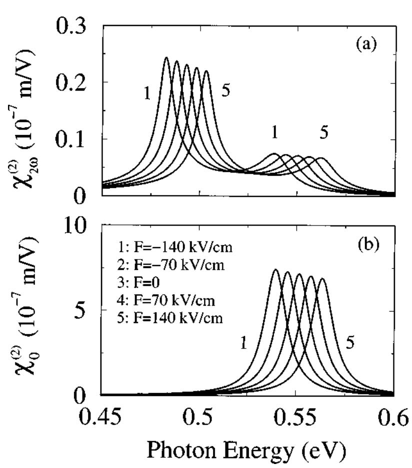

FIG. 2. Second-harmonic ($\chi_{2\omega}^{(2)}$) and optical rectification ($\chi_0^{(2)}$) susceptibilities of a GaN/AlN quantum well as a function of the fundamental photon energy for different applied fields. The QW parameters are the same as in Fig. 1. The line broadening is $\Gamma_{kl}=7$ meV.

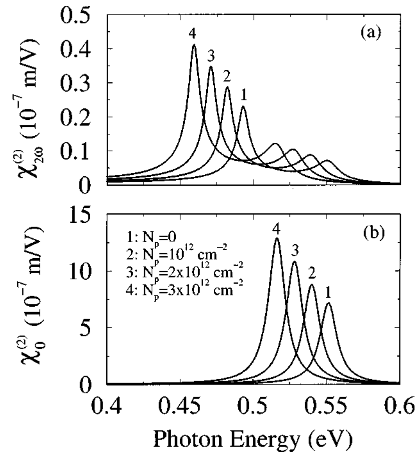

FIG. 3. Second-harmonic ($\chi_{2\omega}^{(2)}$) and optical rectification ($\chi_0^{(2)}$) susceptibilities of a GaN/AlN quantum well as a function of the fundamental photon energy for different interband optical pump-induced electron-hole sheet densities. The QW parameters are the same as those in Fig. 1. The line broadening is $\Gamma_{kl}=7$ meV.

The idea of using the interband optical pump to control the linear intersubband absorption in QWs has been experimentally demonstrated.[14] The interband pump field creates an electron-hole plasma. The photoinduced electrons enhance the intersubband absorption. The many-body screening also changes the single particle energy, so that the absorption peak position is also modified by the electron-hole density. Here we propose to control the *nonlinear* processes in the QW by means of an external optical pump. To calculate the second harmonic and optical rectification susceptibilities with an interband pump, we again solve self-consistently the effective-mass Schrödinger equation and Poisson equations taking into account the fact that the pump field induces a sheet electron-hole density N_p. The exchange-correlation effect is also included. In Fig. 3 we show the second-harmonic ($\chi_{2\omega}^{(2)}$) and optical rectification ($\chi_0^{(2)}$) susceptibilities of a GaN/AlN QW as a function of the fundamental photon energy $\hbar\omega$ for different electron-hole pair densities. With an increase in the electron-hole density, the peak value of the nonlinear coefficients increase as ex-

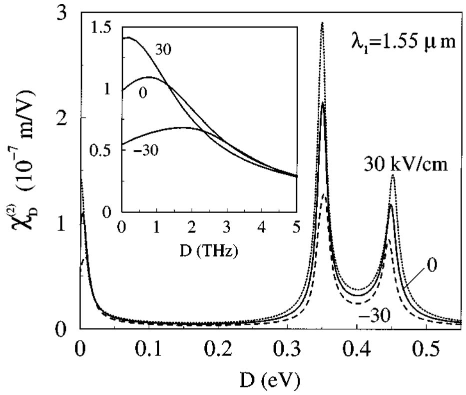

FIG. 4. Difference-frequency generation ($\chi_D^{(2)}$) susceptibility of a GaN/$Al_{0.75}Ga_{0.25}N$ quantum well as a function of the difference frequency $D=\hbar(\omega_1-\omega_2)$ for different applied fields. The fundamental photon energy $\hbar\omega_1=0.8$ eV is fixed. The other QW parameters are the same as those in Fig. 1. The line broadening is $\Gamma_{kl}=7$ meV.

pected. The peak location in the nonlinear spectra is significantly redshifted with an increase of electron-hole density because the plasma screening reduces effectively the quantum size effect so that the intersubband transition energies of the QW become smaller. These results suggest that the second-order nonlinear processes can be easily controlled by the interband pump field.

Here we address the possibility of generating THz radiation in a GaN/$Al_xGa_{1-x}N$ QW structure by using a conventional pump source of 1.55 μm. To this end, we design a symmetric GaN/$Al_{0.75}Ga_{0.25}N$ QW with a GaN well width of 6 nm. For an ionized donor concentration of $5\times10^{18}\,\mathrm{cm}^{-3}$, our calculations show that the intersubband energy $E_{31}=0.795$ eV, which is very close to the 1.55 μm laser frequency. With an applied field we can tune the energy E_{31}, depending upon the sign of the applied field. Fixing one of the fundamental photon energies, $\hbar\omega_1$, at 0.8 eV (1.55 μm), we calculate the difference-frequency generation coefficient by varying the photon energy $\hbar\omega_2$. We see from Fig. 4 that there are three resonances in the nonlinear optical spectra. One is at the difference frequency $D=\hbar(\omega_1-\omega_2)=0$. The other two are located at $D=E_{32}$ and E_{21}, respectively. These resonances are expected because one of the fundamental frequencies is near the transition energy E_{31} so that when the other fundamental frequency hits an intersubband resonance, an enhancement in the nonlinear coefficient is obtained. In the inset of Fig. 4 we expand the frequency around a few THz. By varying the applied field, we change the peak location of the nonlinear coefficient. For example, without the applied field the maximum value appears at $D=0.7$ THz, while at a field of -30 kV/cm the maximum position is near $D=1.8$ THz. To obtain a large $\chi_D^{(2)}$ for the difference-frequency generation in the THz range, it is possible to improve our design such as using coupled quantum wells so that E_{32} is in the THz range. Nevertheless, Fig. 4 shows that the enhancement of $\chi_D^{(2)}$ is about a factor of 2 (compare the peak values at $D=0.3464$ eV and $D=0$). Also, we point out that other QW structures such as InGaAs/AlAsSb,[15] which have a large band edge discontinuity, can also be designed to have the same features with field control or built-in asymmetry, and can be pumped at 1.55 or 1.48 μm wavelengths.

In conclusion, we present a study of second-order nonlinear processes in a wurtzite GaN/AlGaN quantum well. We show that the large strain-induced PZ field results in huge second-order nonlinear susceptibilities (about two-orders of magnitude larger than the bulk value of GaAs). We also investigate the effects of the applied field and the interband optical pump field on the second-order nonlinear processes in the QW. Because of the presence of the PZ field, an applied field can lead to both a blue and red shift of the optical spectrum. The interband pump results in only a redshift of the spectrum because of the plasma screening. Owing to the large band offset and the large PZ field, the GaN/AlGaN QW may find an application in the frequency around 1.55 μm.

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[1] L. Tsang, D. Ahn, and S. L. Chuang, Appl. Phys. Lett. **52**, 697 (1988).
[2] M. M. Fejer, S. J. B. Yoo, R. L. Byer, A. Harwit, and J. S. Harris, Jr., Phys. Rev. Lett. **62**, 1041 (1989).
[3] E. Rosencher, P. Bois, J. Nagle, E. Costard, and S. Delaitre, Appl. Phys. Lett. **55**, 1597 (1989).
[4] E. Rosencher, P. Bois, B. Vinter, J. Nagle, and D. Kaplan, Appl. Phys. Lett. **56**, 1822 (1990).
[5] S. J. B. Yoo, M. M. Fejer, R. L. Byer, and J. S. Harris, Jr., Appl. Phys. Lett. **58**, 1724 (1991).
[6] C. Sirtori, F. Capasso, D. L. Sivco, S. N. G. Chu, and A. Cho, Appl. Phys. Lett. **59**, 2302 (1991).
[7] L. Tsang and S. L. Chuang, Appl. Phys. Lett. **60**, 2543 (1992).
[8] A. Liu, Opt. Commun. **119**, 191 (1995).
[9] A. Liu and O. Keller, Phys. Rev. B **49**, 13616 (1994).
[10] Y. R. Shen, *The Principles of Nonlinear Optics* (Wiley, New York, 1984).
[11] O. Gunnarsson and B. I. Lundqvist, Phys. Rev. B **13**, 4274 (1976).
[12] M. Kumagai, S. L. Chuang, and H. Ando, Phys. Rev. B **57**, 15 303 (1998).
[13] S.-H. Park and S. L. Chuang, Phys. Rev. B **59**, 4725 (1999).
[14] F. H. Julien, in *Intersubband Transitions in Quantum Wells*, edited by E. Rosencher, B. Vinter, and B. Levine (Plenum, New York, 1992), pp. 163–172, and references therein.
[15] A. Neogi, T. Mozume, H. Yoshida, and O. Wada, IEEE Photonics Technol. Lett. **11**, 633 (1999).